

Investigation of Transport Properties for $\text{FeSe}_{1-x}\text{Te}_x$ Thin Films under Magnetic Fields

Yuichi Sawada, Fuyuki Nabeshima, Yoshinori Imai^{*†}, and Atsutaka Maeda

Department of Basic Science, the University of Tokyo, 3-8-1 Komaba, Meguro-ku, Tokyo 153-8902, Japan

We investigated the transport properties under magnetic fields of up to 9 T for $\text{FeSe}_{1-x}\text{Te}_x$ thin films on CaF_2 . Measurements of the temperature dependence of the electrical resistivity revealed that for $x = 0.2 - 0.4$, where T_c is the highest, the width of the superconducting transition increased with increasing magnetic field, while the width was almost the same with increasing magnetic field for $x = 0 - 0.1$. In addition, the temperature dependence of the Hall coefficient drastically changed between $x = 0.1$ and 0.2 at low temperatures. These results indicate that clear differences in the nature of the superconductivity and electronic structure exist between $x = 0 - 0.1$ and $x \geq 0.2$.

The discovery of iron-based superconductors has triggered much attention for fundamental studies and applications.¹ One of the iron-based superconductors, FeSe has the simplest crystal structure, composed of conducting planes alone.² Although the superconducting transition temperature, T_c , of FeSe is 8 K, which is low compared with other iron-based superconductors, T_c reaches as high as 30 K under high pressure.^{3,4} In addition, monolayer FeSe films on SrTiO_3 substrates exhibit very high T_c .⁵ Although it is under debate whether these high T_c have the same origin or not, these results demonstrate that FeSe has potential as a high- T_c superconductor. To raise its T_c , the partial substitution of Te for Se is also effective. $\text{FeSe}_{1-x}\text{Te}_x$ has T_c of up to 14 K at $x = 0.5 - 0.6$.⁶ In addition, the fabrication of thin films makes T_c for $\text{FeSe}_{0.5}\text{Te}_{0.5}$ higher than that of bulk crystals.⁷⁻⁹ Therefore, the fabrication of $\text{FeSe}_{1-x}\text{Te}_x$ thin films is important for both fundamental studies and applications.

It is known that single-phase bulk samples with $0.1 \leq x \leq 0.4$ cannot be obtained because of a phase separation, and this fact has prevented the complete understanding of $\text{FeSe}_{1-x}\text{Te}_x$.⁶ Recently, we have succeeded in obtaining $\text{FeSe}_{1-x}\text{Te}_x$ thin films with these compositions on

[†]Present address: Department of Physics, Tohoku University, 6-3, Aramaki Aza-Aoba, Aoba-ku, Sendai 980-8578, Japan

^{*}imai@tohoku.ac.jp

Table I. Specifications of the $\text{FeSe}_{1-x}\text{Te}_x$ thin films.

x	Thickness (nm)	a (Å)	c (Å)	T_c^{onset} (K)	T_c^{zero} (K)
0	197	3.735	5.584	14.6	13.2
0.1	77	3.753	5.585	11.5	10.1
0.2	41	3.749	5.710	22.6	20.3
0.3	64	3.753	5.784	22.1	20.8
0.4	47	3.758	5.874	21.7	20.5
0.5	78	3.765	5.976	18.3	17.6
0.6	91	3.752	6.066	16.0	15.4
0.7	141	3.755	6.132	13.4	12.9
0.8	148	3.791	6.194	10.4	9.5

CaF_2 substrates by pulsed laser deposition (PLD).¹⁰ T_c for these films increases with decreasing x for $0.2 \leq x \leq 1$ and reaches 23 K at $x = 0.2$. This value is 1.5 times higher than the highest value obtained for bulk samples. Surprisingly, we observed the sudden suppression of T_c between $x = 0.1$ and 0.2. Therefore, it is of great interest to investigate the difference in the physical properties other than T_c in these ranges of x .

In this letter, we will show the temperature dependence of the electrical resistivity under magnetic fields and the Hall effect for $\text{FeSe}_{1-x}\text{Te}_x$ thin films in order to clarify how the transport properties change as a function of x . Our results show that the superconducting transition width, upper critical field, and Hall coefficient change in the range $x = 0.1 - 0.2$, suggesting that a definite change indeed takes place in the electronic structure of $\text{FeSe}_{1-x}\text{Te}_x$ in this range of x .

In this study, all of the films were grown by PLD with a KrF laser. $\text{FeSe}_{1-x}\text{Te}_x$ polycrystalline pellets ($x = 0 - 0.8$) were used as targets.^{11,12} The substrate temperature, laser repetition rate, and base pressure were 280 °C, 20 Hz, and 10^{-7} Torr, respectively. Commercially available $\text{CaF}_2(100)$ substrates, which are one of the most suitable materials for the thin-film growth of $\text{FeSe}_{1-x}\text{Te}_x$,¹²⁻¹⁴ were used for the present experiments. The films were deposited with a six-terminal shape using a metal mask for transport measurements. The measured area was 0.95 mm long and 0.2 mm wide. We measured the temperature dependence of the electrical resistivity and Hall effect for thin films under magnetic fields of up to 9 T by using a Physical Property Measurement System (PPMS, Quantum Design, Inc.). The specifications of the measured films are summarized in Table I. In this table, x is the nominal composition of the polycrystalline target. In a previous paper,¹² we demonstrated that the nominal Te content of the polycrystalline target was nearly identical to that of the final $\text{FeSe}_{1-x}\text{Te}_x$ film from the systematic change in the c -axis length. The lattice parameters shown in Table I, which were

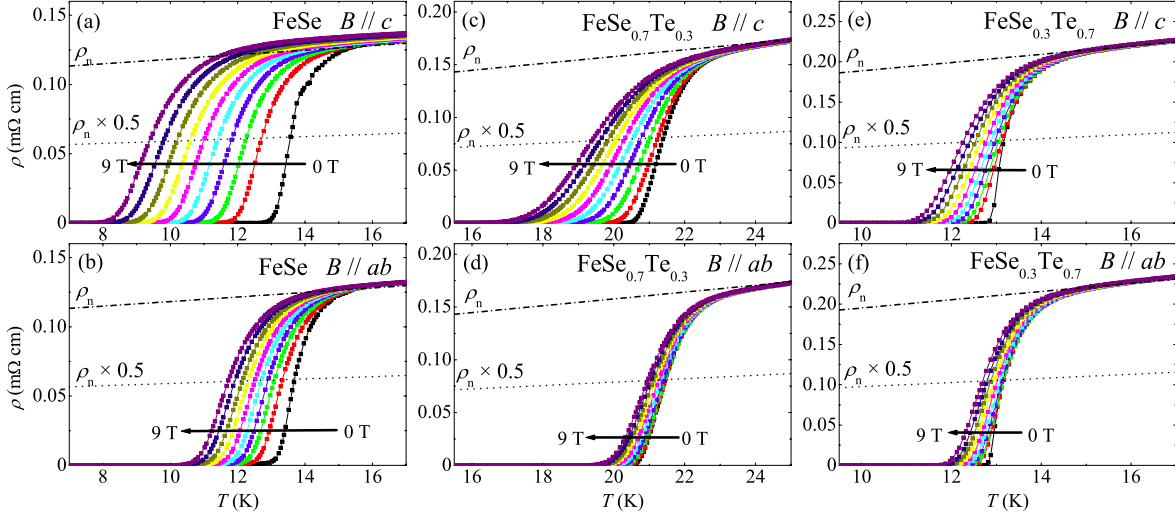


Fig. 1. (Color online) Temperature dependence of the electrical resistivity under magnetic fields B of up to 9 T for $\text{FeSe}_{1-x}\text{Te}_x$ thin films with (a) $x = 0$, $B // c$, (b) $x = 0$, $B // ab$, (c) $x = 0.3$, $B // c$, (d) $x = 0.3$, $B // ab$, (e) $x = 0.7$, $B // c$, and (f) $x = 0.7$, $B // ab$.

estimated from XRD measurements, are almost the same as those reported in our previous paper.¹⁰ Thus, in this paper, we use the nominal value of the Te content of the target as the film composition. The film thicknesses in Table I were measured using a Dektak 6-M stylus profiler. For $\text{FeSe}_{1-x}\text{Te}_x$, it is known that the value of T_c strongly depends on the film thickness.^{7, 10, 14} It should be noted that the optimum film thickness for obtaining the highest T_c depends on the Te composition. Therefore, we controlled the thickness of the measured film such that the highest value of T_c was obtained for each composition. The values of T_c^{onset} and T_c^{zero} were estimated from the temperature dependence of the electrical resistivity.

Figure 1 shows the temperature dependence of the electrical resistivity of $\text{FeSe}_{1-x}\text{Te}_x$ films under finite magnetic fields B of up to 9 T, where the magnetic fields were applied along the ab plane and c -axis. The results are classified into three groups (Groups A, B, and C), in terms of the Te content x . For Group A with $x = 0 - 0.1$, as shown in Figs. 1(a) and 1(b), where T_c is relatively low, the superconducting transition width is almost constant with increasing magnetic field (the so-called parallel shift). This parallel shift is often observed in conventional superconductors. On the other hand, for Group B with $x = 0.2 - 0.4$, as shown in Figs. 1(c) and 1(d), the superconducting transition width increases with increasing magnetic field (so-called resistive broadening), especially for $B // c$. Resistive broadening was also reported by Zhuang *et al.*¹⁵ Finally, for Group C with $x = 0.5 - 1$, as shown in Figs. 1(e) and 1(f), resistive broadening is observed with increasing magnetic field. These results suggest that the nature of superconductivity is different between Group A and Groups B and

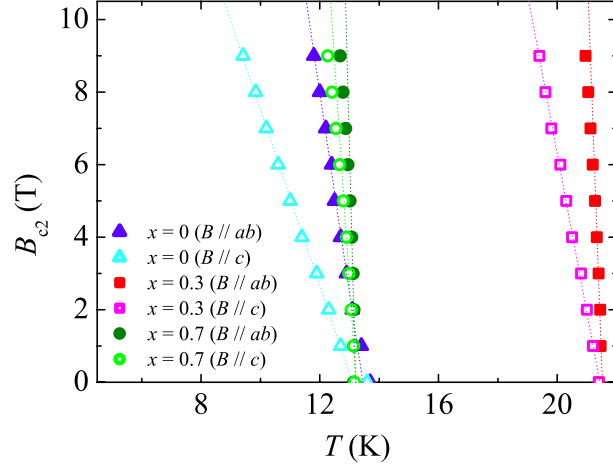


Fig. 2. (Color online) Temperature dependence of B_{c2} along the ab plane and c -axis of $\text{FeSe}_{1-x}\text{Te}_x$ thin films with $x = 0, 0.3$, and 0.7 . Dotted lines are linear fits to the data.

C.

It is important to discuss the origins of resistive broadening for $\text{FeSe}_{1-x}\text{Te}_x$ films with $x \geq 0.2$. Before discussing this case, we recall the origin for high- T_c cuprate, since resistive broadening is familiar in cuprates.¹⁶ The origin is considered to be the result of superconducting fluctuations due to strong two-dimensionality.¹⁷ To examine whether the same scenario as cuprates is applicable for $\text{FeSe}_{1-x}\text{Te}_x$ thin films, we focus on the anisotropy of the upper critical field, $\gamma \equiv B_{c2,0K}^{//ab} / B_{c2,0K}^{//c}$. Figure 2 shows the temperature dependence of the upper critical field B_{c2} along the ab plane and c -axis for the films with $x = 0, 0.3$, and 0.7 . For $\text{FeSe}_{1-x}\text{Te}_x$, the estimation of B_{c2} at 0 K from low-magnetic-field data by utilizing Werthamer–Helfand–Hohenberg (WHH) theory is very difficult because this theory does not take multiband materials into account.¹⁸ For $\text{FeSe}_{1-x}\text{Te}_x$, it is widely accepted that multiple bands, which originate from Fe 3d orbitals, cross the Fermi level.¹⁹ Moreover, the value of B_{c2} at low temperatures is strongly suppressed by the Pauli paramagnetic effect.^{20,21} However, in order to compare B_{c2} for each x within the orbital limit, we consider that the orbital limit inferred using conventional WHH theory is a first-step barometer in the discussion, and we estimate B_{c2} at 0 K using conventional WHH theory. Figure 3(a) shows the x dependence of B_{c2} at 0 K along the ab plane and c -axis. As well as T_c for these films, the value of B_{c2} drastically changes between $x = 0.1$ and 0.2 . The value of B_{c2} for $x = 0.2$ is more than twice that for $x = 0.1$.

Using these values, we estimate the anisotropy of the upper critical field γ . Figure 3(b) shows the x dependence of γ . The value of γ is 1.5 – 3 and not so different between $x = 0.1$

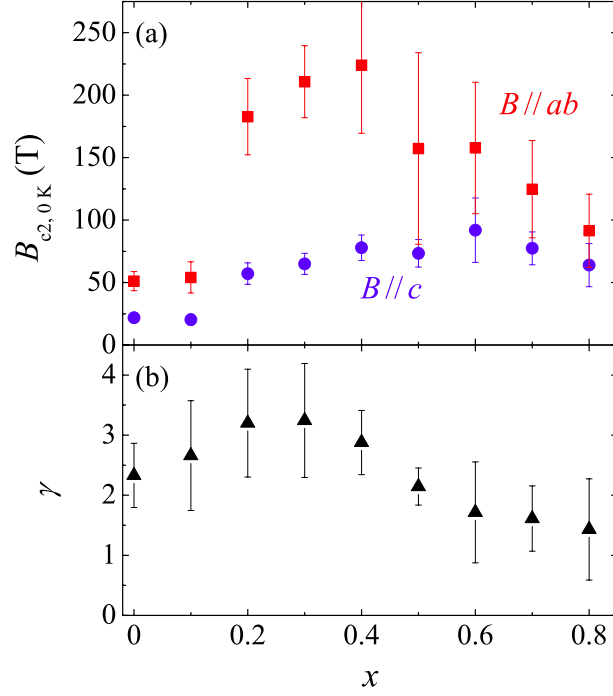


Fig. 3. (Color online) (a) x dependence of B_{c2} for $\text{FeSe}_{1-x}\text{Te}_x$ thin films along the ab plane and c -axis at 0 K estimated from WHH theory within the orbital limit.¹⁸ (b) x dependence of anisotropy $\gamma \equiv B_{c2,0K}^{//ab} / B_{c2,0K}^{//c}$.

and 0.2, in spite of the large difference in T_c and B_{c2} , indicating that the origin of resistive broadening for cuprates is not applicable for $\text{FeSe}_{1-x}\text{Te}_x$ thin films. In the early stages of the research on cuprates, not only superconducting fluctuations but also models of vortex motion across current lines, such as the giant-flux-creep model,²² Kosterlitz–Thouless transition model,²³ and vortex-glass model,²⁴ were proposed as possible origins of resistive broadening. From the above experiments, it is obvious that the nature of superconductivity changes between $x = 0.1$ and 0.2, and further experiments are needed in order to clarify the origin of the resistive broadening.²⁵

Next, we show the results of the Hall effect for $\text{FeSe}_{1-x}\text{Te}_x$ thin films. Figure 4 shows the magnetic field dependence of the Hall resistivity for $\text{FeSe}_{1-x}\text{Te}_x$ thin films. At the lowest temperatures, the resistivity for $x = 0.2$ and 0.3 shows nonlinear behavior as a function of the applied magnetic field. This behavior is the result of the multiband structure for $\text{FeSe}_{1-x}\text{Te}_x$.^{19,26,27} To be precise, we should take all of the bands into account. However, we adopt a two-carrier model including one electron-type carrier (with electron density n_e and mobility μ_e) and one hole-type carrier (with hole density n_h and mobility μ_h) for simplicity. Using this model, the Hall coefficient R_H , which is the slope of the Hall resistivity in the

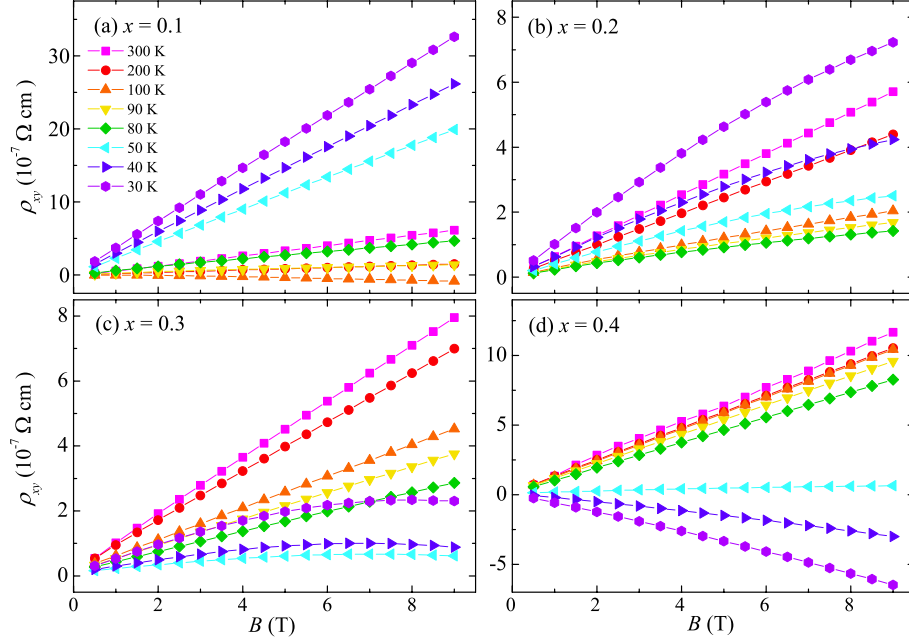


Fig. 4. (Color online) Magnetic field dependence of Hall resistivity for $\text{FeSe}_{1-x}\text{Te}_x$ thin films with (a) $x = 0.1$, (b) $x = 0.2$, (c) $x = 0.3$, and (d) $x = 0.4$.

low-magnetic-field limit, can be expressed as

$$R_H = e^{-1}(n_h\mu_h^2 - n_e\mu_e^2)/(n_h\mu_h + n_e\mu_e)^2. \quad (1)$$

Figure 5 shows the temperature dependence of R_H for $\text{FeSe}_{1-x}\text{Te}_x$ thin films with $x = 0 - 0.5$. At room temperature, the sign of R_H is positive for all films. Above 100 K, R_H for $x = 0$ and 0.1 decreases as the temperature decreases, and below 100 K, it starts to increase rapidly. These results indicate that hole-type transport is dominant at low temperatures. The increase in R_H may be related to the nematicity in FeSe .^{28–30} In contrast, it has been reported that the sign of R_H for FeSe single crystals is negative at low temperatures.^{31,32} The origin of the difference in R_H can be explained by the difference in the band structures between single crystals and films on CaF_2 . Recent angle-resolved photoemission spectroscopy measurements show that the Dirac points of FeSe single crystals are situated in the vicinity of E_F . However, the Dirac points of FeSe films on CaF_2 are at some distance from E_F , compared with that for FeSe single crystals.³³ The difference in the band structures may lead to the difference in R_H between single crystals and films on CaF_2 at low temperatures.

For the $\text{FeSe}_{1-x}\text{Te}_x$ films with $0.2 \leq x \leq 0.5$, which show large values of T_c , the sudden increase in R_H is not observed below 100 K, and the values of R_H are about zero at the lowest temperature. From Eq. (1), these behaviors of R_H indicate that the mobilities of the hole-type and electron-type carriers are comparable at the lowest temperatures. Previously, we

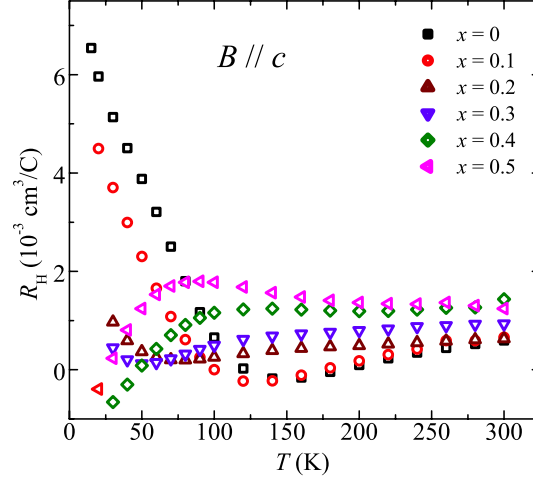


Fig. 5. (Color online) Temperature dependence of Hall coefficient for $\text{FeSe}_{1-x}\text{Te}_x$ thin films with $x = 0 - 0.5$.

reported the temperature dependence of R_H for films with $x = 0.5$,²⁶ and we proposed that T_c strongly depends on the mobility of both electron-type and hole-type carriers. Judging from the behavior of R_H for $\text{FeSe}_{1-x}\text{Te}_x$ thin films with $x = 0 - 0.5$ shown in Fig. 5, a higher T_c is obtained when the mobilities of hole-type and electron-type carriers are comparable. This is consistent with our previous proposal.²⁶ The different behavior of $R_H(T)$ for $x \leq 0.1$ and $x \geq 0.2$ is in good agreement with the dependence of T_c on x . As was pointed out before, the sudden increase in R_H below 100 K in films with $x = 0 - 0.1$ is likely the result of the change in the electronic structure derived from the nematic transition. Thus, our results suggest that the suppression of T_c for $x < 0.1$ is due to the electronic nematicity. In order to further clarify the origin of the suppression of T_c , it is important to measure the Hall resistivity under higher magnetic fields, the results of which will be discussed in a separate publication.

In conclusion, we have investigated the temperature dependence of the electrical resistivity under magnetic fields and Hall resistivity in $\text{FeSe}_{1-x}\text{Te}_x$ thin films. As well as the suppression of T_c between $x = 0.1$ and 0.2 , the superconducting transition width under a magnetic field, B_{c2} , and the low-temperature behavior of the Hall coefficient change between $x = 0.1$ and 0.2 . Our results indicate that the sudden suppression of T_c in the range $x = 0.1 - 0.2$ is closely related to the changes in the nature of the superconductivity and electronic structure.

Acknowledgments

We would like to thank Masafumi Hanawa at CRIEPI (Central Research Institute of Electric Power Industry) for his support in the estimation of the thickness of our films and Kazunori Ueno (Department of Basic Science, the University of Tokyo) for providing the X-ray instrument. This work was supported by JSPS KAKENHI Grant Numbers 15K17697

and 26·9315 and the “Nanotechnology Platform” (Project No.12024046) of the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan.

References

- 1) Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *J. Am. Chem. Soc.* **130**, 3296 (2008).
- 2) F. C. Hsu, J. Y. Luo, K. W. Yeh, T. K. Chen, T. W. Huang, P. M. Wu, Y. C. Lee, Y. L. Huang, Y. Y. Chu, D. C. Yan, and M. K. Wu, *Proc. Natl. Acad. Sci. U.S.A.* **105**, 14262 (2008).
- 3) S. Masaki, H. Kotegawa, Y. Hara, H. Tou, K. Murata, Y. Mizuguchi, and Y. Takano, *J. Phys. Soc. Jpn.* **78**, 063704 (2009).
- 4) S. Medvedev, T. M. McQueen, I. A. Troyan, T. Palasyuk, M. I. Erements, R. J. Cava, S. Naghavi, F. Casper, V. Ksenofontov, G. Wortmann, and C. Felser, *Nat. Mater.* **8**, 630 (2009).
- 5) Q. Y. Wang, Z. Li, W. H. Zhang, Z. C. Zhang, J. S. Zhang, W. Li, H. Ding, Y. B. Ou, P. Deng, K. Chang, J. Wen, C. L. Song, K. He, J. F. Jia, S. H. Ji, Y. Y. Wang, L. L. Wang, X. Chen, X. C. Ma, and Q. K. Xue, *Chin. Phys. Lett.* **29**, 037402 (2012).
- 6) M. H. Fang, H. M. Pham, B. Qian, T. J. Liu, E. K. Vehstedt, Y. Liu, L. Spinu, and Z. Q. Mao, *Phys. Rev. B* **78**, 224503 (2008).
- 7) E. Bellingeri, I. Pallecchi, R. Buzio, A. Gerbi, D. Marrè, M. R. Cimberle, M. Tropeano, M. Putti, A. Palenzona, and C. Ferdeghini, *Appl. Phys. Lett.* **96**, 102512 (2010).
- 8) K. Iida, J. Hänisch, M. Schulze, S. Aswartham, S. Wurmehl, B. Büchner, L. Schultz, and B. Holzapfel, *Appl. Phys. Lett.* **99**, 202503 (2011).
- 9) I. Tsukada, M. Hanawa, T. Akiike, F. Nabeshima, Y. Imai, A. Ichinose, S. Komiya, T. Hikage, T. Kawaguchi, H. Ikuta, and A. Maeda, *Appl. Phys. Express* **4**, 053101 (2010).
- 10) Y. Imai, Y. Sawada, F. Nabeshima, and A. Maeda, *Proc. Natl. Acad. Sci. U.S.A.* **112**, 1937 (2015).
- 11) Y. Imai, R. Tanaka, T. Akiike, M. Hanawa, I. Tsukada, and A. Maeda, *Jpn. J. Appl. Phys.* **49**, 023101 (2010).
- 12) Y. Imai, T. Akiike, M. Hanawa, I. Tsukada, A. Ichinose, A. Maeda, T. Hikage, T. Kawaguchi, and H. Ikuta, *Appl. Phys. Express* **3**, 043102 (2010).
- 13) M. Hanawa, A. Ichinose, S. Komiya, I. Tsukada, T. Akiike, Y. Imai, T. Hikage, T. Kawaguchi, H. Ikuta, and A. Maeda, *Jpn. J. Appl. Phys.* **51**, 010104 (2012).
- 14) F. Nabeshima, Y. Imai, M. Hanawa, I. Tsukada, and A. Maeda, *Appl. Phys. Lett.* **103**, 172602 (2013).

- 15) J. Zhuang, W. K. Yeoh, X. Cui, X. Xu, Y. Du, Z. Shi, S. P. Ringer, X. Wang, and S. X. Dou, *Sci. Rep.* **4**, 7273 (2014).
- 16) Y. Iye, T. Tamegai, H. Takeya, and H. Takei, *Jpn. J. Appl. Phys.* **26**, L1057 (1987).
- 17) R. Ikeda, T. Ohmi, and T. Tsuneto, *J. Phys. Soc. Jpn.* **60**, 1051 (1991).
- 18) N. R. Werthamer, E. Helfand, and P. C. Hohenberg, *Phys. Rev.* **147**, 296 (1966).
- 19) A. Subedi, L. Zhang, D. J. Singh, and M. H. Du, *Phys. Rev. B* **78**, 134514 (2008).
- 20) T. Kida, M. Kontani, Y. Mizuguchi, Y. Takano, and M. Hagiwara, *J. Phys. Soc. Jpn.* **79**, 074706 (2010).
- 21) H. Lei, R. Hu, E. S. Choi, J. B. Warren, and C. Petrovic, *Phys. Rev. B* **81**, 094518 (2010).
- 22) M. Tinkham, *Phys. Rev. Lett.* **61**, 1658 (1988).
- 23) S. Martin, A. T. Fiory, R. M. Fleming, G. P. Espinosa, and A. S. Cooper, *Phys. Rev. Lett.* **62**, 677 (1989).
- 24) K. A. Müller, M. Takashige, and J. G. Bednorz, *Phys. Rev. Lett.* **58**, 1143 (1987).
- 25) K. Kitazawa, S. Kambe, M. Naito, I. Tanaka, and H. Kojima, *Jpn. J. Appl. Phys.* **28**, L555 (1989).
- 26) I. Tsukada, M. Hanawa, S. Komiya, T. Akiike, R. Tanaka, Y. Imai, and A. Maeda, *Phys. Rev. B* **81**, 054515 (2010).
- 27) K. K. Hyunh, Y. Tanabe, T. Urata, H. Oguro, S. Heguri, K. Watanabe, and K. Tanigaki, *Phys. Rev. B* **90**, 144516 (2014).
- 28) K. Horigane, H. Hiraka, and K. Ohoyama, *J. Phys. Soc. Jpn.* **78**, 074718 (2009).
- 29) K. Nakayama, Y. Miyata, G. N. Phan, T. Sato, Y. Tanabe, T. Urata, K. Tanigaki, and T. Takahashi, *Phys. Rev. Lett.* **113**, 237001 (2014).
- 30) A. Maeda, F. Nabeshima, H. Takahashi, T. Okada, Y. Imai, I. Tsukada, M. Hanawa, S. Komiya, and A. Ichinose, *Appl. Surf. Sci.* **312**, 43 (2014).
- 31) M. D. Watson, T. Yamashita, S. Kasahara, W. Knafo, M. Nardone, J. Béard, F. Hardy, A. McCollam, A. Narayanan, S. F. Blake, T. Wolf, A. A. Haghihirad, C. Meingast, A. J. Schofield, H. v. Löhneysen, Y. Matsuda, A. I. Coldea, and T. Shibauchi, *Phys. Rev. Lett.* **115**, 027006 (2015).
- 32) Y. Sun, S. Pyon, and T. Tamegai, *Phys. Rev. B* **93**, 104502 (2016).
- 33) G. N. Phan, K. Nakayama, K. Sugawara, T. Sato, T. Urata, Y. Tanabe, K. Tanigaki, F. Nabeshima, Y. Imai, A. Maeda, and T. Takahashi, private communication.